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(54) Abstract Title
Acrylic resin composition for moulding artificial marble articles

(57) An acrylic resin composition for including an artificial marble article having a curved portion contains 0.1 - 2.5 parts of paraffin by weight per 100 parts of acrylic resin.
The composition is press moulded e.g. to form a washbowl in a counter top.

lubricant.

page 5. amount of additives
page 4. paraffin: lubricant
page 7: syrup of acrylic
page 6: mould release agent

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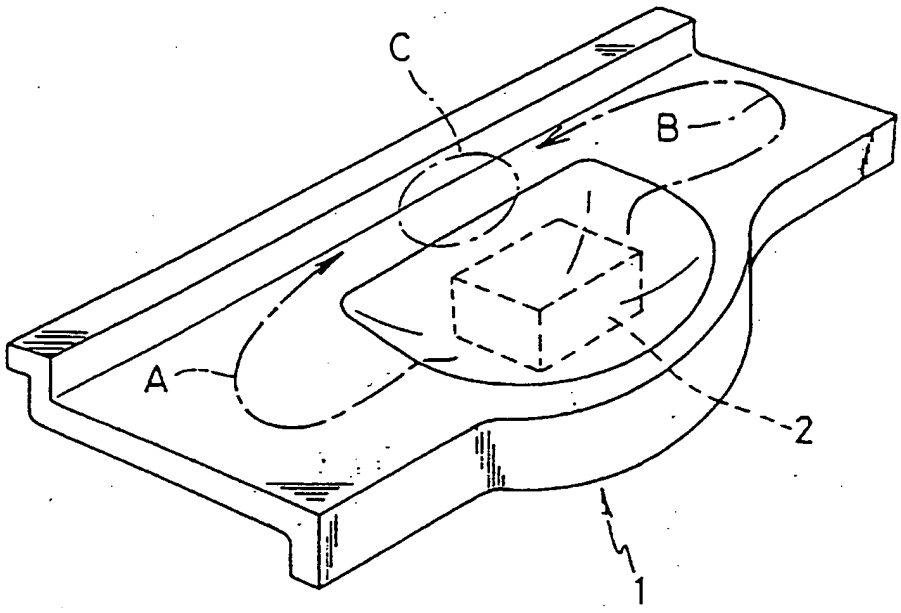
FIG.1



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Acrylic Resin Compound for Artificial Marble and Method of Molding Acrylic Artificial Marble Article

The present invention relates to an acrylic resin compound capable of providing artificial marble of which surface characteristics are excellent and thus having improved appearance, and a method of molding an acrylic artificial marble article by press molding.

To manufacture artificial marble from an acrylic resin compound, casting is mainly employed in the present state of the art. However, press molding offers following advantages over casting. Thus, the deployment of technology in manufacturing artificial marble by press molding is anticipated.

1. The press molding can offer shortened molding cycle for mass production. That is, the press molding can be conducted in short molding cycle of 50% through 20% as compared to the casting, in case of the mass production of more than 20 tons per month.

2. The press molding is conducted at higher temperatures and pressures as compared to the casting, thereby providing higher hardness in molded articles and thus providing more stable hard articles.

As mentioned above, the press molding can offer the faster molding cycle than the casting so that it is better for the mass production. In addition, the articles are cured at high temperatures and high pressures, thereby stabilizing the qualities of the articles and providing good working conditions. However, the finish of the molded

surface of an article molded by the press molding may strongly depend on the configuration of the mold. Therefore, depending on the configuration of the mold, i.e. the configuration of the product, the press molding is likely to provide an article of which surface characteristics is not good.

For example, to mold a counter top with washbowl 1 as shown in Fig. 1 from an acrylic resin compound by the press molding, a material (BMC compound) 2 is generally disposed at a position shown by broken lines in Fig. 1 and then molded by the press molding. In this case, the material 2 flow in such two ways as shown by arrows A, B so that the heads of flows meet each other at a position C where the molded article may have a disorderly surface.

Though the detailed cause of the molding defect in the meeting position C has not been apparent, it is considered that the cause may be vaporization of monomer molecules at the heads of flows of the material, orientation of fibers in the compound due to flowing, decreasing in the uniformity of the compound due to flowing, poor in fluidity of the material at the molding temperature, and so on.

The article having such a disorderly surface has a poor appearance, thereby destroy the high-grade atmosphere, the advantage of the artificial marble. In case of notable case, the articles may not be for sale.

It is an aim of the present invention to provide an acrylic resin compound for artificial marble capable of providing artificial marble, of which surface characteristics are excellent and thus having improved appearance, by press molding regardless of the configuration of a mold, and a method of molding an acrylic artificial marble article from the compound by press molding.

It is another aim of the present invention to provide an acrylic resin compound for artificial marble capable of providing artificial marble, of which surface

characteristics are excellent and thus having improved appearance, by press molding, and a method of molding an acrylic artificial marble article with curves by press molding.

An acrylic resin compound for artificial marble according to a first aspect of the present invention is used for manufacturing an artificial marble by press molding and is characterized by containing one kind or plural kinds of additives selected from a group including silicone oil, surface-active agent, coupling agent, reactive monomer with high boiling point, and reactive oligomer with high boiling point.

A method of molding an acrylic artificial marble article according to a second aspect of the present invention is characterized in that the aforementioned acrylic resin compound for artificial marble is molded by press molding.

That is, close examination of the inventors for preventing the occurrence of a disorderly surface at the meeting position where heads of flows of the material meet each other in the conventional press molding indicated that the molding defect can be prevented by adding one kind or plural kinds of additives selected from a group including silicone oil, surface-active agent, coupling agent, reactive monomer with high boiling point, and reactive oligomer with high boiling point into the acrylic resin compound, wherein each additive has

(a) a boiling point higher than the molding temperature,

(b) compatibility or homogeneous dispersion against vinyl monomer mainly consisting of methyl methacrylate composing the acrylic resin, and

(c) low surface tension.

The effects of the present invention due to such additives may be as follows.

The high boiling point of the additive prevents the vaporization of monomer molecules from the material. The low surface tension of the additive improves the fluidity of the materials against the cavity face because the additive oozes out of the surface of the flowing material. It also improves the drape between fibers and filler

and matrix resin, thereby preventing the failure of evenness of the material and thus preventing the defect at the meeting position of the heads of flows.

An acrylic resin compound according to a third aspect of the present invention is used for molding an acrylic artificial marble article with curves and is characterized by containing 0.1 - 2.5 parts of paraffin by weight per 100 parts of acrylic resin.

A method of molding an acrylic artificial marble article according to a fourth aspect of the present invention is characterized in that an acrylic artificial marble article with curves is molded from the aforementioned acrylic resin compound by press molding.

That is, close examination of the inventors for preventing the occurrence of a disorderly surface at the meeting position where heads of flows of the material meet each other in the conventional press molding indicated that the molding defect can be prevented by adding a given amount of paraffin into the acrylic resin compound.

The effects of the present invention due to the paraffin may be as follows.

Since the paraffin has high boiling point, the vaporization of monomer molecules from the material can be prevented. The paraffin oozes out of surface of the flowing material so as to function as lubricant, thereby improving the fluidity of the materials against the cavity face and preventing the oxygen hardening.

Preferred embodiments of the present invention will now be described hereinbelow by way of example only with reference to the accompanying drawings, in which:

Fig. 1 is a perspective view showing a counter top with washbowl manufactured in accordance with the preferred embodiments of the present invention and comparative examples.

Hereinafter, the present invention will be described in detail.

First, the description will be made as regard to any acrylic resin compound for

artificial marble according to the first embodiment of the present invention and a method of molding acrylic artificial marble article made of the compound according to the second embodiment of the present invention.

According to the first and second embodiments, to prevent the molding defect in the meeting position where heads of flows of the material meet each other during the process of the press molding, one kind or plural kinds of additives selected from a group including silicone oil, surface-active agent, coupling agent, reactive monomer with high boiling point, and reactive oligomer with high boiling point are added in the acrylic resin compound.

Little amounts of additives to be added may not produce a sufficient effect, while large amounts of additives may destroy the characteristics, the advantage of acrylic resin compound for artificial marble, since the amounts of the other compositions are relatively reduced.

In case of adding silicone oil, the amount of the silicone oil to be added is preferably from 0.1 % to 10 % by weight for the acrylic resin. In case of adding surface-active agent, the amount of the surface-active agent to be added is preferably from 0.1 % to 5 % by weight for the acrylic resin. In case of adding coupling agent, the amount to be added is preferably from 0.1 % to 5 % by weight for the acrylic resin. In case of adding reactive monomer with high boiling point, the amount to be added is preferably from 1 % to 30 % by weight for the acrylic resin. In case of adding reactive oligomer with high boiling point, the amount to be added is preferably from 1 % to 30 % by weight for the acrylic resin.

It should be noted that the surface-active agent may be N-methylpyrrolidone soap, and the coupling agent may be an agent having functional lipophilic group and three hydrophilic groups, such as vinyl-silane or vinyl-trimethoxy-silane.

It should be also noted that the reactive monomer with high boiling point may have the boiling point higher than the molding temperature, i.e. more than 140°C.

Therefore, for example, polymerized monomer, such as styrene or caprolactam, may be employed as the reactive monomer with high boiling point.

Further, the reactive oligomer with high boiling point may have the boiling point higher than the molding temperature, i.e. more than 140°C. Therefore, for example, acrylic oligomer, such as polysiloxane containing acrylic group, or oligomer of multifunctional acrylate may be employed as the reactive oligomer with high boiling point.

The acrylic resin compound for artificial marble according to the first embodiment may be the same composition as the conventional acrylic resin compound but the additive as mentioned above to be added. The acrylic resin compound may be the sum of acrylic resin compound, generally consisting of the following composition, plus the aforementioned additive.

The composition of the acrylic resin compound besides the aforementioned additive

Acrylic resin : 100

Filler : 100 - 350

Reinforcing fiber : 0 - 30

Curing agent : 0.05 - 3.0

Internal mold release agent : 0 -50

It should be noted that the filler may be calcium carbonate, barium sulfate, talc, silica, alumina, aluminum hydroxide, or glass ball. Particularly, aluminum hydroxide having excellent workability may be preferably employed for artificial marble to manufacture a counter top or the like, while silica having good water resisting property and surface hardness may be preferably employed for artificial marble to manufacture a bathtub or the like. The reinforcing fiber may be glass fiber, carbon fiber, or organic fiber or the like. The curing agent may be one belonging to peroxy ester group, such as t-butylperoxy-2-ethylhexanoate and the internal mold release agent may be zinc stearate or calcium stearate.

Besides the composition as mentioned above, the acrylic resin compound for artificial marbie according to the first embodiment may contain thickening agent such as magnesium oxide, polymerization inhibitor such as methyl tertiary butyl hydroquinone, ultraviolet absorber (UV absorber), anti-foaming agent, or/and finishing agent.

It should be noted that the acrylic resin may be acrylic syrup containing: vinyl monomer mainly consisting of methyl methacrylate (MMA) : 40 - 70 % by weight, crosslinking agent : 0 - 10 % by weight, and methyl methacrylate polymer having average molecular weight of 10,000 - 150,000 : 30 - 60 % by weight, wherein the crosslinking agent is preferably multifunctional acrylate such as dimethacrylate-1,6-hexanediol.

As mentioned above, the vinyl monomer mainly consists of methyl methacrylate. The ratio of the methyl methacrylate in the vinyl monomer is equal to or more than 50 % by weight, preferably more than 75 % by weight, because methyl methacrylate of less than 50 % of weight of the vinyl monomer reduces the weatherability and the stain resistance of the resultant artificial marble.

The vinyl monomer which can be used in combination with the methyl methacrylate may be methacrylate ester besides the methyl methacrylate, acrylic ester, or aromatic vinyl monomer. The methyl methacrylate polymer (MMA polymer) may contain carboxyl group.

The acrylic resin compound for artificial marble according to the first embodiment is effectively available particularly for molding an article having an intricate configuration which facilitates occurrence of a disorderly surface at the meeting position where heads of flows of the material meet each other during the process of the press molding, such as a counter top with washbowl, a counter top with sink of a kitchen unit, or the like.

An artificial marble article can be manufactured from the acrylic resin compound of the first embodiment as mentioned above by press-molding the acrylic resin

compound at the molding temperature of 105 - 140°C with the temperature difference of 10 - 20 °C between the surface and the back of the article, and at the molding pressure of 3 - 10 MPa in the conventional method.

With the acrylic resin compound for artificial marble of the first embodiment and the method of molding acrylic artificial marble article made of the compound according to the second embodiment of the present invention, an artificial marble of which surface characteristics are excellent and thus having improved appearance can be obtained by press molding regardless of the configuration of a mold.

The description will be made as regard to acrylic resin compound for artificial marble according to the third embodiment of the present invention and a method of molding acrylic artificial marble article made of the compound according to the fourth embodiment of the present invention.

According to the third and fourth embodiments, to prevent the molding defect in the meeting position where heads of flows of the material meet each other during the process of the press molding, paraffin is added in the acrylic resin compound.

When the amount of the paraffin is less than 0.05 parts by weight per 100 parts of acrylic resin, there is no prospect of a sufficient effect, while the paraffin of more than 2.5 parts by weight may destroy the characteristics in strength and the like, the advantage of acrylic resin compound for artificial marble, since the amounts of the other composition are relatively reduced.

The paraffin is preferably paraffin wax with melting point of 40 - 60°C since the appearance of the resultant product is good.

The acrylic resin compound for artificial marble according to the third embodiment may be the same composition as the conventional acrylic resin compound but the paraffin as mentioned above to be added. Generally, the acrylic resin compound may be the sum of the acrylic resin compound, consisting of the same as stated in the description of the acrylic resin compound for artificial marble according to the first

embodiment, plus the paraffin between 0.1 and 2.5 parts by weight.

In the same manner of the first embodiment, besides the aforementioned composition, the acrylic resin compound for artificial marble according to the third embodiment may contain thickening agent such as magnesium oxide, polymerization inhibitor such as methyl-t-butyl hydroquinone, UV absorber, anti-foaming agent, or/and finishing agent.

It should be noted that the acrylic resin may be acrylic syrup containing: vinyl monomer mainly consisting of methyl methacrylate (MMA) : equal to or more than 50 % by weight, crosslinking agent : 0 - 10 % by weight, and methyl methacrylate polymer having average molecular weight of 10,000 - 150,000 : equal to or less than 50 % by weight, wherein the crosslinking agent is preferably multifunctional acrylate such as dimethacrylate-1,6-hexanediol.

As mentioned above, the vinyl monomer mainly consists of methyl methacrylate. The ratio of the methyl methacrylate in the vinyl monomer is equal to or more than 50 % by weight, preferably more than 75 % by weight, because methyl methacrylate of less than 50 % of weight of the vinyl monomer reduces the weatherability and the stain resistance of the resultant artificial marble.

The vinyl monomer which can be used in combination with the methyl methacrylate may be methacrylate ester besides the methyl methacrylate, acrylic ester, or aromatic vinyl monomer. The methyl methacrylate polymer (MMA polymer) may contain carboxyl group.

The acrylic resin compound for artificial marble according to the third embodiment is effectively available particularly for molding an article having an intricate deep-drawing configuration which facilitates occurrence of a disorderly surface at the meeting position where heads of flows of the material meet each other during the process of the press molding, such as a counter top with washbowl, a counter top with sink of a kitchen unit, or the like.

An artificial marble article with curves can be manufactured from the acrylic resin compound of the third embodiment as mentioned above by press-molding the acrylic resin compound with a mold for press molding at the molding temperature of 105 - 140°C with the temperature difference of 10 - 20 °C between the surface and the back of the article, and at the molding pressure of 3 - 10 MPa.

With the acrylic resin compound for artificial marble of the third embodiment and the method of molding acrylic artificial marble article made of the compound according to the fourth embodiment of the present invention, an artificial marble with curves of which surface characteristics are excellent and thus having improved appearance can be obtained by press molding.

Hereinafter, the present invention will now be described in further detail with reference to preferred embodiments and comparative examples.

Embodiments 1 - 5, Comparative Example 1

A counter top with washbowl 1 as shown in Fig. 1 was manufactured with each acrylic resin compound as prepared stated in Table 1 under the molding condition as follows. It should be noted that the material 2 is disposed at a position shown by broken lines in Fig. 1. The compositions of the acrylic resin are as follows.

Molding Condition

Molding Temperature	Temperature of the surface of the mold : 130°C
	Temperature of the back of the mold : 120°C
Molding Pressure	5 MPa
Molding Period	8 minuets

Composition of the Acrylic Resin (% by weight)

MMA monomer : 60

Crosslinking Agent (multifunctional acrylate) : 1

MMA polymer (the average molecular weight 120,000) : 39

[Table 1]

	Examples	Embodiments					C. Ex.
		1	2	3	4	5	1
Composition of Material (parts by weight)	Acrylic Resin	100	100	100	100	100	100
	Aluminum Hydroxide	200	200	200	200	200	200
	Glass Fiber	10	10	10	10	10	10
	Zinc Stearate	1	1	1	1	1	1
	Curing Agent	1	1	1	1	1	1
	Silicone Oil	2	0	0	0	0	0
	Caprolactam	0	5	0	0	0	0
	N-methylpyrrolidone	0	0	2	0	0	0
	Vinyl-trimethoxy-silane	0	0	0	2	0	0
	Polysiloxane with acrylic group	0	0	0	0	5	0

C. Ex = Comparative Example, Curing Agent = Peroxide

As a result, there are appearance defect due to the disorderly surface at the meeting position C in Fig. 1 in the comparative example 1. On the other hand, there are no disorderly surface in either the embodiment 1 containing the silicone oil, the embodiment 2 containing the caprolactam, the embodiment 3 containing the N-methylpyrrolidone, the embodiment 4 containing the vinyl-trimethoxy-silane, or the embodiment 5 containing Polysiloxane with acrylic group, thereby making the resultant articles to have good appearance.

Embodiments 6 - 10, Comparative Examples 2 - 4

The counter top with washbowl 1 as shown in Fig. 1 was manufactured with each acrylic resin compound as prepared stated in Table 2 under the molding condition

as follows. It should be noted that the material 2 is disposed at the position shown by broken lines in Fig. 1. The compositions of the acrylic resin are as follows.

Molding Condition

Molding Temperature	Temperature of the surface of the mold : 130°C
	Temperature of the back of the mold : 120°C
Molding Pressure	5 MPa
Molding Period	8 minutes

Composition of the Acrylic Resin (% by weight)

MMA monomer : 60

Crosslinking Agent (multifunctional acrylate) : 1

MMA polymer (the average molecular weight 100,000) : 39

The appearance of each resultant article was observed and the strength (Falling Ball Impact Strength: Hmax, the maximum of height H causing no crack when falling a ball of 200g from an upper position Hmm to the article) was also observed. Table 2 shows the results of the observation.

As apparent from Table 2, an article having improved appearance and high strength can be manufactured by adding paraffin of 0.1 - 2.5 parts by weight per 100 parts of acrylic resin.

[Table 2]

Examples		C. Ex.		Embodiments					C.Ex
		2	3	6	7	8	9	10	4
Composition of Material ; parts by weight	Acrylic Resin	100	100	100	100	100	100	100	100
	Filler	150	150	150	150	150	150	150	150
	Reinforcing fiber	10	10	10	10	10	10	10	10
	Internal Mold Release Agent	5	5	5	5	5	5	5	5
	Curing Agent	1.2	1.2	1.2	1.2	1.2	1.2	1.2	1.2
	Polymerization Inhibitor	0.1	0.1	0.1	0.1	0.1	0.1	0.1	0.1
	Coupling Agent	1.0	1.0	1.0	1.0	1.0	1.0	1.0	1.0
	Thickening Agent	1.0	1.0	1.0	1.0	1.0	1.0	1.0	1.0
	Paraffin (Melting point: 53°C)	0	0.05	0.1	0.5	1.0	2.0	2.5	3.0
Appearance *		NG1	NG1	Good	Good	Good	Good	Good	NG2
Strength	Evaluation	○	○	○	○	○	○	○	×
	Hmax	100	100	100	100	100	100	100	90

C. Ex = Comparative Example, Filler = Aluminum Hydroxide, Reinforcing Fiber = Glass Fiber : Fiber length of 1.5 mm, Internal Mold Release Agent = Zinc Stearate, Curing Agent = Peroxide, Polymerization Inhibitor = Methyl-t-butyl hydroquinone, Thickening Agent = Magnesium Oxide

* NG1 : disorder appeared at end.

NG2 : unevenness of gloss appeared on surface.

Good : Excellent. Neither disorderly nor unevenness appeared.

Embodiments 11 - 20, Comparative Examples 5 -10

These are molded in the same manner as the embodiments 6 - 10 and the comparative examples 2 - 4 but paraffin having melting point of 43°C or 69°C. Then the appearance and the strength were observed. Tables 3, 4 show the results.

As compared with the Tables 2 through 4, it was found that the paraffin having melting point between 40°C and 60°C was preferable, and particularly among the three

kinds of employed paraffin, the paraffin having melting point of 53 °C was the best.

[Table 3]

Melting point of Paraffin : 69°C

Examples		C. Ex.		Embodiments					C.Ex
		5	6	11	12	13	14	15	7
Parts of Paraffin by weight		0	0.05	0.1	0.5	1.0	2.0	2.5	3.0
Appearance *		NG1	NG1	Good	Good	Good	Good	Good	NG2
Strength	Evaluation	○	○	○	○	○	○	○	×
	Hmax	100	100	100	100	100	100	100	90

* NG1 : disorder appeared at end.

NG2 : unevenness of gloss appeared on surface.

Good : Excellent. Neither disorderly nor unevenness appeared.

[Table 4]

Melting point of Paraffin : 43°C

Examples		C. Ex.		Embodiments					C.Ex
		8	9	16	17	18	19	20	10
Parts of Paraffin by weight		0	0.05	0.1	0.5	1.0	2.0	2.5	3.0
Appearance *		NG1	NG1	Good	Good	Good	Good	Good	NG2
Strength	Evaluation	○	○	○	○	○	○	○	×
	Hmax	100	100	100	100	100	100	100	90

* NG1 : disorder appeared at end.

NG2 : unevenness of gloss appeared on surface.

Good : Excellent. Neither disorderly nor unevenness appeared.

CLAIMS:

1. An acrylic resin compound for molding an acrylic artificial marble article having a curved portion, wherein the acrylic resin compound contains 0.1 - 2.5 parts of paraffin by weight per 100 parts of acrylic resin.

2. An acrylic resin compound as claimed in claim 1, wherein said paraffin has melting point of 40 - 60 °C.

3. An acrylic resin compound as claimed in claim 1, wherein said compound comprises 100 parts of acrylic resin by weight, 100 - 350 parts of filler by weight, 0 - 30 parts of reinforcing fiber by weight, 0.05 - 3.0 parts of curing agent by weight, 0 - 50 parts of internal mold release agent by weight, and 0.1 - 2.5 parts of paraffin by weight.

4. An acrylic resin compound as claimed in claim 3, wherein the filler is at least one of aluminium hydroxide and silica, and the reinforcing fiber is glass fiber.

5. An acrylic resin compound as claimed in claim 3, wherein the acryiic resin is acrylic syrup comprising: not less than 50% vinyl monomer mainly consisting of methyl methacrylate by weight, 0 - 10% crosslinking agent by weight, and not more than 50% methyl methacrylate polymer, having average molecular weight of 10,000 - 150,000, by weight.

6. An acrylic resin compound as claimed in claim 5, wherein the ratio of methyl methacrylate in the vinyl monomer mainly consisting of methyl methacrylate is equal to or more than 75% by weight.

7. An acrylic resin compound as claimed in claim 5, wherein the crosslinking agent is multifunctional acrylate.

8. An acrylic resin compound as claimed in claim 3 wherein the filler is glass.

9. An acrylic resin compound as claimed in claim 8 wherein the glass is glass balls.

10. A method of molding an acrylic artificial marble article from an acrylic resin compound for artificial marble by press molding, wherein the acrylic resin compound contains 0.1 - 2.5 parts of paraffin by weight per 100 parts of acrylic resin.

11. A method of molding an acrylic artificial marble article as claimed in claim 10, wherein the press molding is conducted at the molding temperature of 105 - 140 °C with the temperature difference of 10 - 20 °C between the surface and the back of the article, and at the molding pressure of 3 - 10 MPa.

12. An acrylic resin compound for manufacturing artificial marble by press molding substantially as hereinbefore described with reference to the accompanying drawing.

13. An acrylic resin compound for manufacturing artificial marble by press molding substantially as hereinbefore described with reference to any of Embodiments 6 to 20.

14. A method of molding an acrylic artificial marble article from an acrylic resin compound for artificial marble by press molding substantially as hereinbefore described with reference to the accompanying drawing.

15. A method of molding an acrylic artificial marble article from an acrylic resin compound for artificial marble by press molding substantially as hereinbefore described with reference to any of Embodiments 6 to 20.

The Patent Office

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Application No:	GB 9908802.3	**Examiner:**	Miss M M Kelman
Claims searched:	1 to 15	**Date of search:**	12 May 1999

Patents Act 1977
Search Report under Section 17

Databases searched:

UK Patent Office collections, including GB, EP, WO & US patent specifications, in:

UK Cl (Ed.Q): C3K KFA KLA, C3M MXC, C3V VBE VDT

Int Cl (Ed.6): C04B 26/06; C08K 5/01, 13/02; C08L 33/10, 33/12

Other: ONLINE: EPODOC, JAPIO, WPI

Documents considered to be relevant:

Category	Identity of document and relevant passage		Relevant to claims
X	GB 1037904 A	Du PONT see the Examples	1,2,3,5,6, 7
X	GB 0975587 A	CHEMISCHE FABRIK GRUNAU see Examples 1 and 2	1,2,3,4,5, 6
A	EP 0318325 A2	NIPPON SHOKUBAI	
A	WPI Abstract Accession No. 94-299944[37] & JP 060228403 A (MITSUBISHI RAYON) 16.08 1994 see abstract		

X	Document indicating lack of novelty or inventive step	A	Document indicating technological background and/or state of the art.
Y	Document indicating lack of inventive step if combined with one or more other documents of same category.	P	Document published on or after the declared priority date but before the filing date of this invention.
&	Member of the same patent family	E	Patent document published on or after, but with priority date earlier than, the filing date of this application.